

An irreversible magnetic-field dependence of low-temperature heat transport of spin-ice compound $\text{Dy}_2\text{Ti}_2\text{O}_7$ in a $[111]$ field

C. Fan,¹ Z. Y. Zhao,¹ H. D. Zhou,^{2,3} X. M. Wang,¹ Q. J. Li,¹ F. B. Zhang,¹ X. Zhao,⁴ and X. F. Sun^{1,*}

¹*Hefei National Laboratory for Physical Sciences at Microscale,*

University of Science and Technology of China, Hefei, Anhui 230026, People's Republic of China

²*Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996-1200, USA*

³*National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32306-4005, USA*

⁴*School of Physical Sciences, University of Science and Technology of China, Hefei, Anhui 230026, People's Republic of China*

(Dated: February 27, 2013)

We study the low-temperature thermal conductivity (κ) of $\text{Dy}_2\text{Ti}_2\text{O}_7$ along and perpendicular to the (111) plane and under the magnetic field along the $[111]$ direction. Besides the step-like decreases of κ at the field-induced transitions from the spin-ice state to the kagomé-ice state and then to the polarized state, an abnormal phenomenon is that the $\kappa(H)$ isotherms show a clear irreversibility at very low temperatures upon sweeping magnetic field up and down. This phenomenon surprisingly has no correspondence with the well-known magnetization hysteresis. Possible origins for this irreversibility are discussed; in particular, a pinning effect of magnetic monopoles in spin ice compound by the weak disorders is proposed.

PACS numbers: 66.70.-f, 75.47.-m, 75.50.-y

I. INTRODUCTION

The isolated magnetic charges or the magnetic monopoles, in contrast to their electrical counterparts, have not been observed in nature. A recent discovery was that the magnetic monopoles could emerge from the collective excitations of spin ice.^{1–10} Although this kind of magnetic-monopole excitations is not completely the same as the free elementary particles, this finding is interesting and very important in the sense that it was the first time to signify the magnetic monopoles in real space.

$\text{Dy}_2\text{Ti}_2\text{O}_7$ (DTO), as a spin ice material, has already become the focus of condensed matter physics¹ and received a lot of attentions during past ten years due to its fantastic physical properties.^{11–19} DTO belongs to the family of pyrochlore titanates with the Dy^{3+} spins locating at the vertexes of tetrahedra, which consist of triangular and kagomé planes stacked alternately along the $[111]$ direction. At low temperatures, the Dy^{3+} spins are Ising anisotropic and form a macroscopically degenerated “2-in, 2-out” spin-ice ground state, which is attributed to the frustration effect between the nearest-neighboring antiferromagnetic interaction and the ferromagnetic dipolar interaction.^{12–14} Once the flipping of a spin occurs, a local “3-in, 1-out” or “3-out, 1-in” spin configuration forms, which is equivalent to yielding two opposite magnetic monopoles in the adjacent tetrahedra,^{16–18} as shown in Fig. 1(a). The monopole pairs can be separated and diffuse freely in the spin-ice state, which can be viewed as a “vacuum” free of charges by continuous spin flip and behaves a random walk process in zero field.^{1,4} A particular case of interest is with the magnetic field along the three-fold axis or the $[111]$ direction. With increasing field, there are two successive transitions from the spin-ice state to the kagomé-ice state and then to the fully-polarized state.^{16–19} In the kagomé-ice state, the mag-

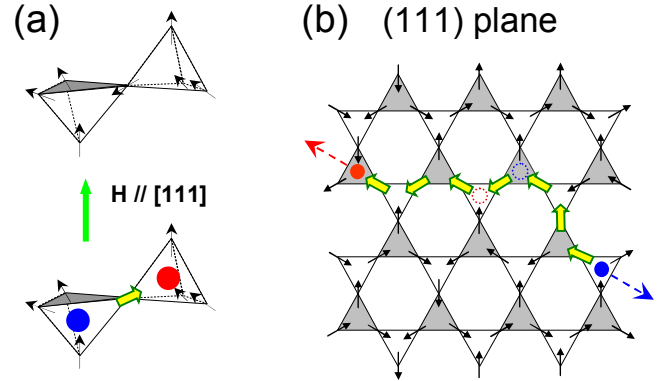


FIG. 1: (Color online) (a) “2-in, 2-out” spin configuration of the spin-ice ground state in $\text{Dy}_2\text{Ti}_2\text{O}_7$. The spin-easy axis is along the three-fold axis or the $[111]$ direction. Two neighboring tetrahedra are shown and the arrows stand for Dy^{3+} spins on the vertexes. When one spin is flipped, indicated by the open arrow, by thermal fluctuations or external magnetic field, a pair of magnetic monopoles are formed and shown by the red and blue dots. (b) In magnetic field perpendicular to the (111) plane, the magnetic monopoles can move in the plane by successive spin flipping. The dashed red and blue arrows, accompanied with the dashed open circles, show the routines of positive and negative monopoles.

netic field pins the spins on the triangular planes along the field direction. However, the ice rule can still be satisfied in low magnetic field. As a result, a reduced degeneracy is expected for the kagomé plane and the dimensionality of the spin system is reduced. Therefore, the magnetic monopoles can diffuse only in the kagomé plane,³ as shown in Fig. 1(b). However, it should be noted that the experimental investigations on probing the monopole excitations have not yet arrived perfect consistency.^{20,21} Studying spin-ice materials by using more different tech-

niques is probably an effective way.

Low-temperature heat transport is a powerful tool to probe the properties of elementary excitations^{22–29} and the magnetic-field-induced magnetic transitions^{30–38}. In principle, the magnetic monopoles, as the elementary excitations in the spin-ice compounds, can also contribute to the heat transport by acting as either heat carriers or phonon scatterers. In this work, we study the low- T thermal conductivity (κ) of DTO single crystals down to 0.3 K with heat current parallel and perpendicular to the (111) plane and in the magnetic field along the [111] axis. It is found that the phonon heat transport shows step-like decreases at the magnetic phase transitions, which is nearly isotropic on the direction of heat current and is mainly due to the magnetic scattering on phonons. Besides, a remarkable hysteresis of $\kappa(H)$ isotherm appears at very low temperatures. The peculiarity is that this irreversibility has no direct correspondence with the well-known magnetization irreversibility. The possible mechanisms are discussed.

II. EXPERIMENTS

High-quality DTO single crystals were grown using the floating-zone technique. The thermal conductivity at low temperatures down to 0.3 K was measured using a conventional steady-state technique.^{33,34,36,37} In this work, we measured thermal conductivities along and perpendicular to the (111) plane, named as κ_{\parallel} and κ_{\perp} , by using two samples with sizes of $5.3 \times 0.74 \times 0.16 \text{ mm}^3$ and $3.1 \times 0.71 \times 0.15 \text{ mm}^3$, respectively. Note that the heat currents have to be along the longest dimension, while the magnetic field is always perpendicular to the (111) plane and is along the shortest and the longest dimension for κ_{\parallel} and κ_{\perp} samples, respectively. So the demagnetization effect is unavoidably different for these two samples. The typical misalignment of the magnetic field is less than 2° .

It is necessary to point out that in this work the magnetic-field dependencies of κ were measured in a static field mode. That is, the measurements were done by the following steps: (i) change field slowly to a particular value and keep it stable; (2) after the sample temperature is stabilized, apply a heat power at the free end of sample; (iii) wait *long enough* time until the temperature gradient on the sample (judged from the time dependencies of two RuO₂ thermometers on the samples) is completely stabilized; (iv) record the temperatures of two thermometers and obtain the temperature gradient. The purpose of this operation is to avoid any misleading data. It is known that DTO has very slow spin dynamics at low temperatures,³⁹ which may cause some relaxation phenomenon in the temperature gradient measurements, if the magnetic excitations are involved in the heat transport behaviors. Actually, in such case, a thermal conductivity measurement in the sweeping-field mode could be incorrect because it is impossible to get a steady heat flow.

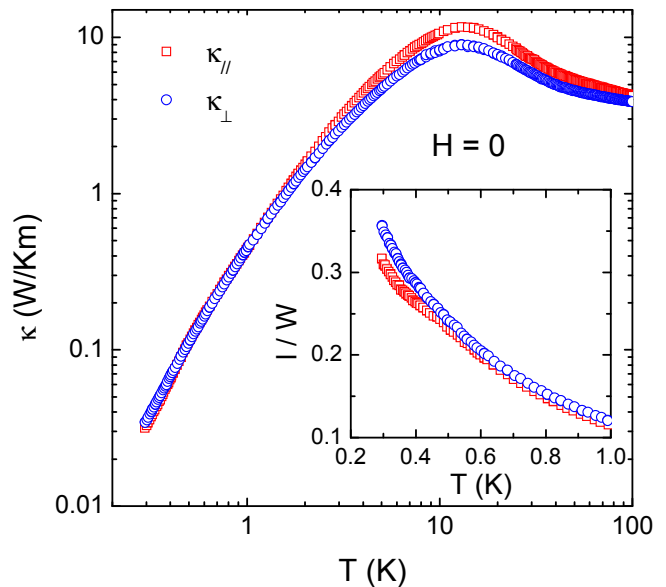


FIG. 2: (Color online) Temperature dependencies of the thermal conductivities of Dy₂Ti₂O₇ in zero field for heat current along the (111) plane or perpendicular to it. The inset shows the temperature dependencies of the phonon mean free path l divided by the averaged sample width W for two sets of data.

III. RESULTS AND DISCUSSION

A. Heat carriers

Figure 2 shows the temperature dependencies of κ_{\parallel} and κ_{\perp} in zero field. Both κ_{\parallel} and κ_{\perp} exhibit clear phonon peaks at ~ 15 K and a rough $T^{2.7}$ dependence at sub-Kelvin temperatures, which is however weaker than the standard T^3 behavior of phonon thermal conductivity at the boundary scattering limit.²² Similar result has been obtained for heat current along the [110] direction, and the magnetic scattering on phonons was discussed to be important at temperatures below 10 K.⁴⁰ It is possible to estimate the mean free path of phonons at low temperatures and to judge whether the phonons are free from microscopic scattering at subKelvin temperatures. The phononic thermal conductivity can be expressed by the kinetic formula $\kappa_{ph} = \frac{1}{3} C v_p l$,²² where $C = \beta T^3$ is the phonon specific heat at low temperatures, v_p is the average velocity and l is the mean free path of phonons. Using the β value obtained from specific-heat measurements (not shown here), the phonon velocity can be calculated and then the mean free path is obtained from the κ .^{36,41} The inset to Fig. 2 shows the ratios l/W for the two samples, where W is the averaged sample width.^{36,41} It is clear that both ratios increase quickly at very low temperatures and are expected to approach 1 below 0.3 K. This estimation tells us that it is not likely that the magnetic excitations in DTO (for examples, magnetic monopoles) can make a sizable contribution to transporting heat, since the experimental κ is even smaller than

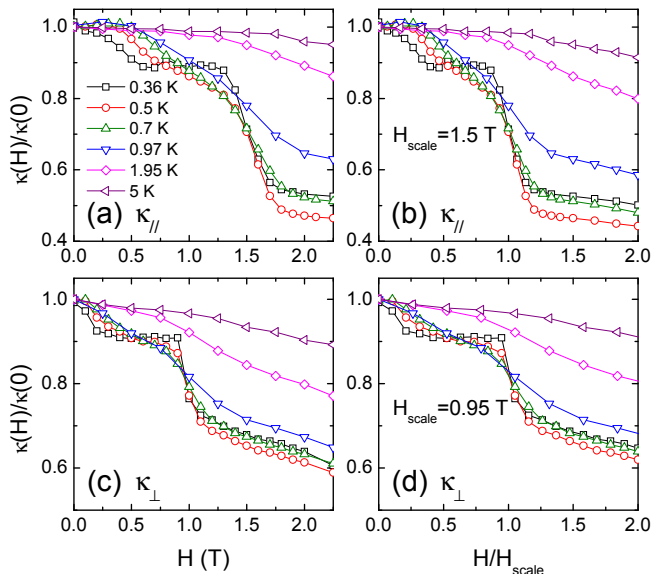


FIG. 3: (Color online) Magnetic-field dependencies of the $\kappa_{||}$ and κ_{\perp} for field along the [111] direction. The data are taken with decreasing field from high-field limit (14 T). Panels (a) and (c) show the raw data, while (b) and (d) show the data with magnetic fields re-scaled by 1.5 and 0.95 T, respectively, considering the demagnetization effect. Note that the data after this re-scaling are essentially isotropic.

the phonon term in a boundary scattering limit. It should be pointed out that the magnetic-monopole excitations were believed to be negligible at very low temperatures, because the energy barrier for flipping a spin or creating a pair of magnetic monopoles is about 4.4 K.⁴²

B. $\kappa(H)$ and field-induced magnetic transitions

Figures 3(a) and 3(c) show the $\kappa(H)$ isotherms for $\kappa_{||}$ and κ_{\perp} , respectively, in magnetic field perpendicular to the (111) plane. Note that these data are taken with sweeping field down from 14 T. One can easily notice that at very low temperatures $\kappa_{||}(H)$ and $\kappa_{\perp}(H)$ display two anomalies at low magnetic fields. For example, at 0.36 K the $\kappa_{\perp}(H)$ curve shows two clear step-like transitions at about 0.25 and 0.9 T, which are in good correspondence with the critical fields of the subsequential transitions from the low-field spin-ice phase to the kagomé-ice phase then to the saturated state (“3-in, 1-out” or “3-out, 1-in”).^{16–18} In present work, we defined these two transition fields as H_{c1} and H_{c2} . The sharpness of the transitions is quickly smeared out upon increasing temperature. Similar results are observed in the low- T $\kappa_{||}(H)$ isotherms. It is clearly seen that the main difference between the $\kappa_{||}$ and κ_{\perp} is that the critical fields of the step-like transitions are somewhat different. However, if the horizontal axis of the $\kappa_{||}(H)$ and $\kappa_{\perp}(H)$ curves are re-scaled by some particular values, as shown in Figs. 3(b) and 3(d), one can obtain the essentially same behavior of $\kappa_{||}$ and κ_{\perp} .

This means that the difference between the experimental data of $\kappa_{||}$ and κ_{\perp} is simply due to the demagnetization effect.¹⁰ Therefore, the heat transport behaviors shown in Fig. 3 actually indicate a nearly isotropic behavior and a close relationship between the heat transport and the magnetic phase transitions.

The drastic change of heat transport at the magnetic phase transition has been widely observed in many kinds of magnetic materials and is closely associated with the evolution of magnetic excitations.^{30–38} However, the particular features of $\kappa(H)$ at the magnetic transitions can be significantly different from each other, which depends on both the natures of magnetic transitions and the roles of magnetic excitations in the heat transport (as heat carriers or phonon scatterers). In principle, the magnetic monopoles as a kind of excitations in the spin-ice and kagomé-ice states can either transport heat or scatter phonons. However, it is easy to know that the magnetic monopoles are not likely to act as heat carriers for two reasons. First, as mentioned above, in zero field the monopole excitations are actually very difficult to emerge at subKelvin temperatures. Second, a low field along the [111] direction can significantly enhance the excitations of magnetic monopoles, whereas the thermal conductivity does not show any increase. Then, can the step-like decreases of κ at two transitions be attributed to phonon scattering by the sudden increases of magnetic monopoles? There is no doubt that the magnetic monopoles can effectively scatter phonons by absorbing phonon energy and flipping spins, and suppress the phonon heat conductivity. However, above H_{c2} (~ 1 –1.5 T), the magnetic monopoles are so populated that they fully occupy the kagomé network with the monopole pairs sitting on the nearest sites, in which case their ability of scattering phonons must be negligible. Therefore, even if the step-like decrease of $\kappa(H)$ at H_{c1} can be explained by the enhanced phonon scattering by magnetic monopoles, the one at H_{c2} cannot be related to the magnetic monopoles.

It is known that, due to the strong Ising anisotropy of Dy^{3+} spins, the magnetic monopoles connected by the Dirac strings are the elementary magnetic excitations of the spin ice state. The $\kappa(H)$ behaviors shown in Fig. 3, however, indicate that there may be other magnetic excitations playing an important role in the step-like decreases of κ at two magnetic transitions. In this regard, there has been no obvious evidence that DTO could have other magnetic excitations than the magnetic monopoles, like the spin fluctuations caused by non-Ising terms in the Hamiltonian.^{21,43} Nevertheless, some drastic field-induced changes related to crystal lattice and phonons have been indeed found. In a recent work, the ultrasound measurements also indicated sharp anomalies of the sound velocity and sound attenuation at H_{c2} , which may share a common origin with the $\kappa(H)$ transitions.¹⁰ It is also notable that a local maximum of entropy at H_{c2} may also indicate a strong spin fluctuations that can strongly scatter phonons.⁴⁴

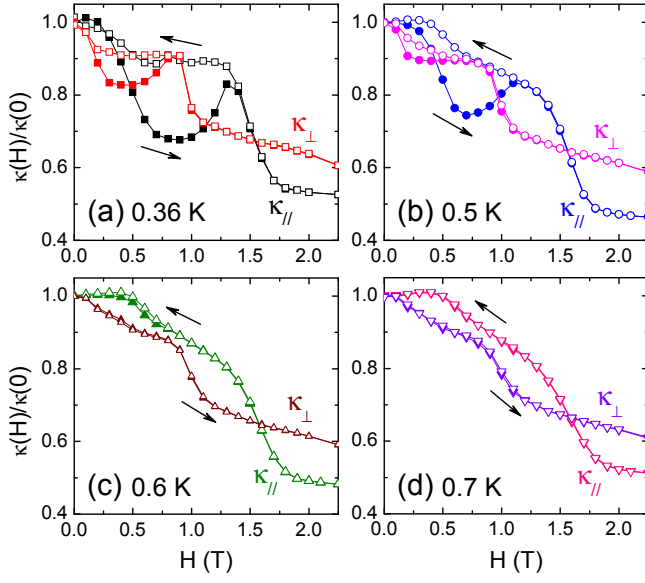


FIG. 4: (Color online) $\kappa_{\parallel}(H)$ and $\kappa_{\perp}(H)$ loops with magnetic field sweeping up and down (perpendicular to the (111) plane). The data shown with solid symbols are measured in ascending field after the sample is cooled in zero field, while the open symbols show the data with descending field, as indicated by arrows. The demagnetization effects, which are the same as those in Fig. 3, are not taken into account in these plots.

Another important impact on the field dependence of κ that should be taken into account is the paramagnetic scattering effect on phonons.^{22,45,46} It is known that the Dy^{3+} ions have a degenerate doublet of the lowest crystal-field level, which was evidenced as a simple two-level Schottky anomaly of the low- T specific heat in magnetic fields.^{47,48} Therefore, a Zeeman-effect splitting of the doublet can produce resonant scattering on phonons and give a low-field suppression of thermal conductivity, as some other magnetic materials have shown.^{45,46} This can also explain the continuous decrease of κ at relatively high temperatures (above ~ 1 K), where two magnetic transitions are gone.

C. Irreversibility of $\kappa(H)$ curves

An unusual phenomenon in DTO is that, as shown in Fig. 4, the $\kappa_{\parallel}(H)$ and $\kappa_{\perp}(H)$ isotherms show an irreversibility at very low temperatures. For example, after cooling the sample to 0.36 K in zero field, both κ_{\parallel} and κ_{\perp} are firstly measured with increasing field, for which they show a valley-and-peak feature. Then, upon decreasing field from above 2 T, the magnitudes of κ_{\parallel} and κ_{\perp} deviate from those of the field-increasing curves below ~ 1.5 and 1 T, respectively. Actually, the differences between field-up and field-down curves are so large that the valley-and-peak feature of $\kappa(H)$ does not show up at all for decreasing field. Upon increasing temperature, the

irreversibility is quickly diminished and disappears above 0.7 K. The peculiarity of this transport irreversibility is that it has no direct correspondence to that of the low- T magnetization curves $M(H)$,¹⁶ which feature a pronounced hysteresis below 0.5 T and a small one around 0.9 T that were ascribed to the slow dynamics of the spin ice and the first-order phase transition, respectively. Furthermore, some higher-field hysteresis of $M(H)$ was observable only below 0.36 K.¹⁶

Although the irreversibility of thermal conductivity can sometimes be observed at the field-induced first-order phase transitions in many materials, they are usually very small and presented in a narrow vicinity of the critical fields.³⁰ One exception for showing large hysteretic heat transport is the high- T_c superconductors in the mixed state,⁴⁹ for which the hysteretic $\kappa(H)$ behaviors are accompanied with the magnetization irreversibility caused by the vortex pinning. In other words, the difference of $\kappa(H)$ between the field-up curve and the field-down one is directly related to the magnetism, which is reasonable because the density and distribution of the vortices determine the strength of vortex-electron scattering and the electron heat transport.⁵⁰ In contrast, the irreversible $\kappa(H)$ behaviors of DTO can exist even in the field region where the magnetization shows a nearly reversible plateau.¹⁶ In addition, the ultrasound properties also displayed a broad hysteresis, but only at temperatures as low as 0.29 K.¹⁰ Since the heat transport measurement is performed in a static-field mode, in contrast to the recent interesting nonstationary magnetization and ultrasound measurements,^{7,10} it may reveal different physics from other techniques.

It is necessary to point out that the step-like decreases in the low- T $\kappa(H)$ isotherms in some sense are not extraordinary, since similar anomalies have been observed in other magnetic materials^{30,37} and are not difficult to understand. Although the hysteresis of $\kappa(H)$ looks less remarkable than the step-like features in the magnitude, it is actually a more peculiar phenomenon related to the magnetism of the spin-ice materials.

D. Possible origins for the irreversibility of $\kappa(H)$

One possible origin of the irreversible $\kappa(H)$ behaviors is related to field misalignment from the [111] direction, which is known to have important impact on the field-induced transitions.⁵¹ When the field is slightly tilted from the [111] direction due to a misalignment, its component parallel to the (111) plane can break the three-fold degeneracy of the modified ice rule. Accordingly, the kagomé spins tend to align at a temperature that depends on this parallel component. This is known as a Kasteleyn transition.⁵¹ An early entropy measurement indicated that a field tilting larger than 2° can significantly smear out the local entropy peak at the H_{c2} .⁴⁴ If such a misalignment of spins could result in a hysteresis in the entropy of the kagomé-ice state when the

field is swept up and down, the phonon heat transport would show a hysteresis considering the magnetic scattering on phonons. Furthermore, the partial ordering caused by the parallel component of field is likely accompanied with hysteresis because of the slow spin dynamics. Since this kind of hysteresis may appear only in the magnetization component perpendicular to the $[111]$ direction, it cannot directly be observed in the magnetization along the $[111]$ direction. Therefore, it would not be surprising that the $\kappa(H)$ irreversibility has no direct correspondence to the well-known $M(H)$ curves. For these reasons, the field misalignment should be carefully taken into account. However, there are two factors to be considered if one tries to ascribe the irreversibility of $\kappa(H)$ to the field misalignment. First, since both the low- T $\kappa_{\parallel}(H)$ and $\kappa_{\perp}(H)$ isotherms in the present work show rather sharp decreases at H_{c2} , the misalignment in our measurements might be negligibly small. Second, it has been not yet proved that such kind of field misalignment can really lead to some irreversible behavior in the kagomé-ice state.

We propose another possibility with a “pinning” effect of the magnetic monopoles by the static disorders, in analogy to the weak-disorder induced pinning effect on vortices in the superconductors. Considering a simple case that the defect is on the nonmagnetic Ti^{4+} site, the interactions of the Dy^{3+} spins in that tetrahedron would not be the same as others and the flipping of these spins could be more difficult. In other words, a magnetic monopole located at this defect site cannot move away freely. Thus, crystal defects can work as pinning centers of the magnetic monopoles. Figure 5 illustrates the movement process of monopoles considering this kind of pinning effect. In zero field, there are few magnetic monopoles induced by thermal fluctuations (at very low temperatures), as shown in Fig. 5(a). When the field is increasing, some spins are flipped and the magnetic monopoles are created in pairs (see Fig. 5(b)) and diffuse freely in the crystal (see Fig. 5(c)). With increasing the field further, the density of magnetic monopoles increases and they could contribute to scattering phonons when they are moving in the (111) plane; this results in a quick suppression of κ , as shown in Figs. 4(a) and 4(b). Note that in this process, once some magnetic monopoles meet with the defects, they can be pinned. Near the saturation field, there are so many monopoles that they gradually lose their mobility, and the scattering effect on phonons becomes weakened and κ can be somewhat recovered. At the saturated state (the spins are fully polarized at H_{c2}), the magnetic monopoles are well populated so that they fully occupy the kagomé network with the monopole pairs sitting on the nearest sites in Fig. 5(d). At this moment, they completely lose the mobility and the κ reaches the local maximum (the decrease of κ at $H > H_{c2}$ is due to other reasons, as was discussed in section III. B). When the field is decreasing across the saturation field, the density of magnetic monopoles is quickly reduced by annihilation in pairs. Most of remained ones may have

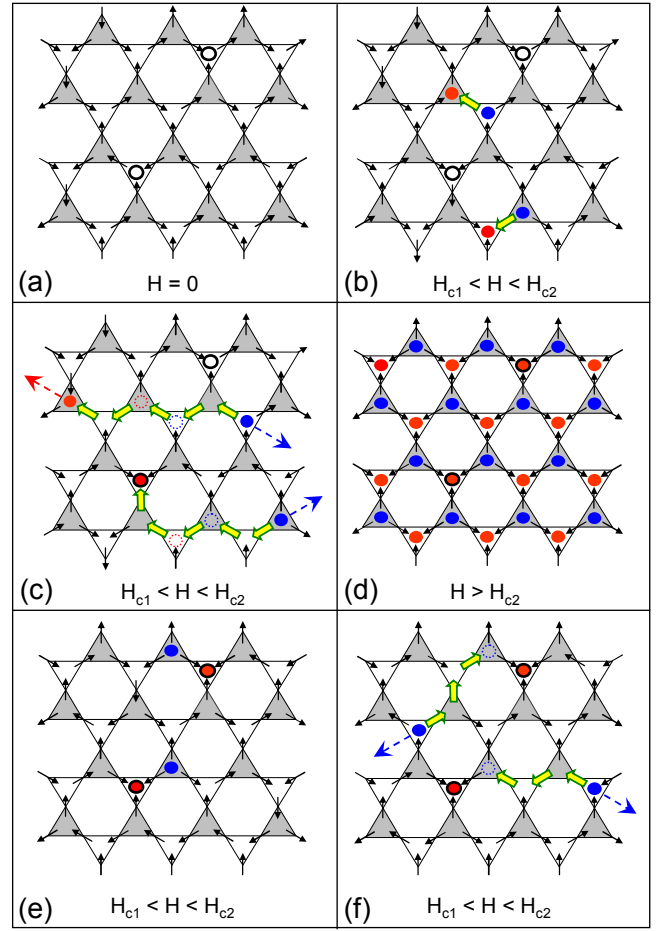


FIG. 5: (Color online) Structure evolution of the spin projection on the (111) plane. The magnetic field is perpendicular to the plane. H_{c1} and H_{c2} are defined in the text. Panels (a)-(d) show the field-up process while (e)-(f) show the field-down process. (a) In zero field and at very low temperatures, the thermally excited magnetic monopoles are very few. The black circles stand for crystal defects. (b) With increasing field, some magnetic monopoles are created in pairs (the red and blue dots). (c) Magnetic monopoles can move separately by spin flipping. The flipped spins are shown by the open and yellow arrows. The dashed red and blue arrows, accompanied with the dashed open circles, show the routines of positive and negative monopoles. Once some magnetic monopoles meet with the defects, they can be pinned. (d) Above H_{c2} , the magnetic monopoles are so well-populated that they cannot move any more. (e) With decreasing field, the number of magnetic monopoles is decreased. (f) Those magnetic monopoles sitting on the defect sites are pinned, while the others still can move in the plane.

some chance to start moving, but those pinned by the defects cannot be mobile, as shown in Figs. 5(e)-(f). As a result, the proportion of the mobile monopoles in the field-decreasing process is smaller than that in the field-increasing case. Consequently, the phonon scattering is weaker in the former case and κ shows an irreversible behavior. It could be expected that the irreversibil-

ity of κ must be quickly weakened with increasing temperature, because the stronger thermal fluctuations will cause the magnetic monopoles to be depinned. One may think that, based on the above discussion, the peculiar heat transport properties can be explained by a simple phonon-assistant spin flip and the magnetic monopoles might be irrelevant. However, it is notable that the irreversibility is larger in $\kappa_{\parallel}(H)$ than that in $\kappa_{\perp}(H)$. This indicates that the magnetic monopoles can more effectively scatter the in-plane phonons as a kind of quasi-particles since they can move in the kagomé plane.³ The validity of this picture needs further experimental and theoretical investigations.

IV. SUMMARY

The very-low-temperature thermal conductivity of $\text{Dy}_2\text{Ti}_2\text{O}_7$ displays an irreversibility with magnetic field applied in the [111] direction, which seems to have no direct correspondence with the magnetization hysteresis.

We discussed possible origins of this irreversibility, including a field misalignment effect and a pinning effect of magnetic monopoles by the weak disorders.

Acknowledgments

We thank Y. Takano for helpful discussions. This work was supported by the National Natural Science Foundation of China, the National Basic Research Program of China (Grant Nos. 2009CB929502 and 2011CBA00111), and the Fundamental Research Funds for the Central Universities (Program No. WK2340000035).

Note added.—We note a recent study on the heat transport of $\text{Dy}_2\text{Ti}_2\text{O}_7$,⁵² in which the measurements were done in magnetic field along the [001] direction. An irreversible behavior of $\kappa(H)$ was also presented in low fields (< 0.3 T), which is directly related to the irreversible magnetization. This is very different from what we observed in a much higher and broader field range.

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- * Electronic address: xfsun@ustc.edu.cn
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